MOLECULAR NANOMAGNETS

Molecular nanomagnets, sometimes referred to as single molecule magnets, are organic materials that contain a very large (Avogadro's) number of magnetic molecules that are (nearly) identical, providing ideal laboratories for the study of nanoscale magnetic phenomena. With molecular clusters of large total spin 10, Mn_{12} and Fe_8 exhibit properties that are borderline between classical and quantum magnetism. They are magnetically bistable at low temperatures, they exhibit "macroscopic quantum tunneling" between up and down spin orientations, and quantum interference between tunneling paths. Interest in these materials has grown dramatically in the last several years, owing to their possible use for high density information storage, as well as the possibility that some member of this family of materials could provide the qubits needed for quantum computation.

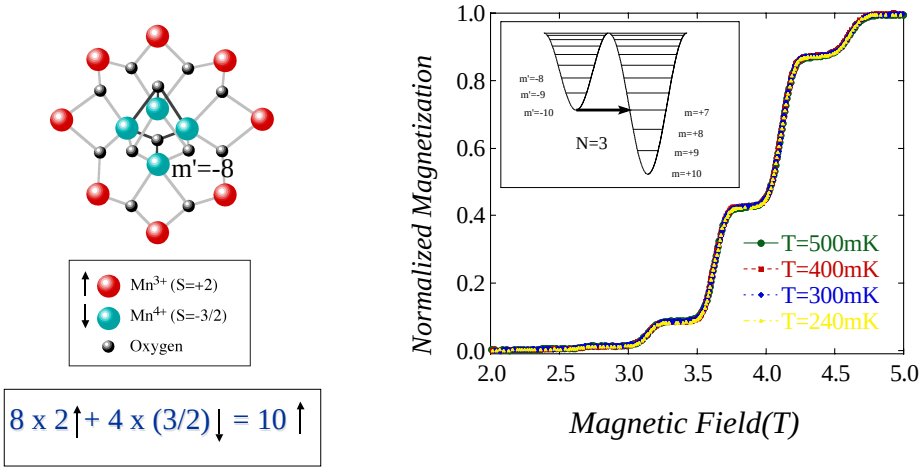

FIG. 1. (a) Schematic of the Mn_{12} molecule composed of four inner spin ($S = -3/2$) Mn^{4+} ions and eight outer spin ($S = +2$) Mn^{3+} ions with oxygen bridges, yielding a total spin $S = 10$ ground state at low temperatures; (b) Magnetization versus longitudinal magnetic field applied along the easy c-axis. The inset shows tunneling for step N=3.

Prototypical of the class, Mn_{12}-Acetate contains magnetic clusters, shown in Fig. 1, composed of twelve Mn atoms coupled by superexchange through oxygen bridges to give a sizable $S = 10$ spin magnetic moment that is stable at temperatures of the order of 10 K and below. The identical weakly-interacting clusters are regularly arranged on a tetragonal crystal lattice. As illustrated by the double well potential of Fig. 1, strong uniaxial anisotropy yields doubly degenerate ground states in zero field and a set of excited levels corresponding to different projections, $m_s = \pm 10, \pm 9, ..., 0$, of the total spin along the easy c-axis of the crystal. Magnetic relaxation proceeds in these systems by spin reversal via thermal excitation over the anisotropy barrier and/or by quantum tunneling across the potential barrier. Below the blocking temperature of ≈ 3 K, a series of steps appear in the curves of M versus H, indicating enhanced relaxation of the magnetization by tunneling whenever levels on opposite sides of the anisotropy barrier coincide in energy.

Various spectroscopic probes have been applied to the study of single molecule magnets. Neutron scattering studies, generally performed in the absence of magnetic field, have been carried out by several groups, as well as studies using NMR techniques. EPR studies of Barra *et al.* on powder samples extend to 525 GHz (17.5 cm^{-1}), while Hill and coworkers span the spectral range from 8 GHz to 250 GHz (0.3 to 8.33 cm^{-1}) in single crystal samples. Studies by Mukhin, Dressel, and coworkers have been carried out from 1.5 to 33 cm^{-1} (45 GHz to 1 THz). Experiments at much high energies by Musfeldt and coworkers encompass the ran obtained in the i

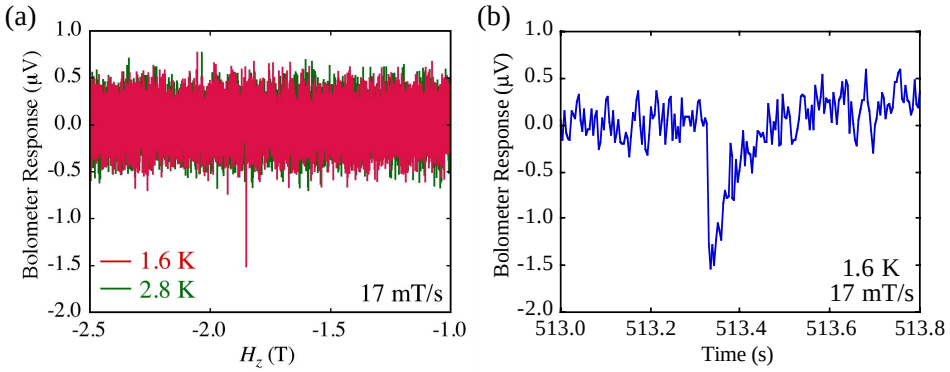

FIG. 2. The bolometer response to the radiation emitted by three Mn_{12} crystal assemblies in a swept magnetic field at 1.6 K and 2.8 K; note the sharp pulse of radiation present at 1.6 K is absent above the blocking temperature at 2.8 K; (b) The measured THz radiation pulse as a function of time showing a decay rate of about 0.1 sec.

Chudnovsky and Garanin [1] have proposed that when the wavelength of the emitted radiation is comparable to or larger than the sample size, single molecule magnets can superradiate, emitting coherent radiation at characteristic frequencies corresponding to transitions between different spin projections along the easy axis. This is an exciting possibility that would be of great interest both from a fundamental viewpoint, as well as the possibility of designing coherent laser sources in the microwave and THz range of the spectrum.

A number of different schemes have been suggested for producing superradiance, the simplest of which is to induce magnetic avalanches which are thought to produce coherent radiation. Using a pumped bolometer that measures photons in the frequency range 30 GHz to 1 THz, we have made preliminary measurements (shown below) at Brookhaven on an assembly of three Mn_{12}-acetate single crystals glued together. Strong bursts of radiation were observed in a swept magnetic field at fields where avalanches were expected to occur; this radiation was not present above the blocking temperature. No information was obtained regarding the frequency content of the emitted radiation, a necessary step to insure that the radiation emitted is not simply due to sample heating during an avalanche As shown in Fig. 2(b), the duration of the emitted radiation is about 0.1 sec.

[1] E. M. Chudnovsky and D. A. Garanin, Phys. Rev. Lett. **89**, 157201 (2002).